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Smectite to Illite Transformation: Effects of Interlayer Cation, Silica Concentration, Fluid Chemistry, and Time

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Bentonite is a clay-bearing material with highly desirable properties for use as backfill in engineered barrier concepts for nuclear waste isolation in deep geological repositories. This is due to the swelling and sorption properties of smectite minerals in bentonite. However, thermally driven diagenetic processes could eventually transform smectite to illite, thus potentially reducing those favorable properties. The process of illitization is dependent on temperature, pressure, the presence of aqueous K^+, and silica concentration in the system. Understanding the effects of various chemical conditions on smectite stability will enhance predictions of long term barrier functions within the safety case framework for bentonite materials in deep geologic repositories. The investigations presented here emphasize the impact of interlayer cations, Na^+, K^+, NH_4^+ and Cs^+, on the alteration of smectite to illite, along with the influence of time and silica concentration. A comprehensive characterization of starting material and reaction products under given experimental conditions allows tailoring the material to assist in slowing/inhibiting smectite to illite transformation.

Less than 2um particles of natural Na-smectite were exchanged with K^+, NH_4^+ and Cs^+ salt solutions to obtain each form, respectively. Solutions of 1M KCl or DI water and clay with varying water/rock ratios (100, 500, and 1000) were loaded into Parr vessel reactors and heated to 200°C for time periods between 7 days to 112 days. Following the reaction, pH was analyzed, and ICP-OES was used to detect dissolved clay constituents. The solid products were characterized by XRD, BET, XRF, and SEM.

The results suggest that natural and K-exchanged smectite converts to illite within 56 days. The transformation is enhanced by increasing K^+ concentration, reduced silica content, and increasing reaction time; indicated prominently by XRD analysis. Further studies to characterize NH_4- and Cs-smectite reaction products are in progress.

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